## REMARKS

The Amendment and Request for Reconsideration filed on July 20, 2004, cited to a textbook by Ronald Probstein (*see* page 16, of Amendment). The cited portion of this text:

> Ronald Probstein, <u>Physicochemical Hydrodynamics, An Introduction</u>, 2nd ed, John Wiley & Sons, Inc., NY, pp. 131-133 (1994)

is attached herewith for the Examiner's convenience (Exhibit 1).

## CONCLUSION

Applicants assert that the above-identified application is in condition for allowance and respectfully request such action at this time.

If any issues remain outstanding, Applicants respectfully request that the Examiner contact the undersigned at the number provided below to schedule a further interview or to discuss any issues by telephone interview.

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## <u>AUTHORIZATIONS</u>

It is believed that no fee is due for filing this submission. However, the

Commissioner is hereby authorized to charge any additional fees which may be required for

entry of this submission, or credit any overpayment to Deposit Account No. 13-4503, Order No.

<u>2324-7028US1</u>. A DUPLICATE COPY OF THIS SHEET IS ATTACHED.

Respectfully submitted,
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Dated: <u>July 27, 2004</u>          By: _____

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# Physicochemical Hydrodynamics

## An Introduction
## Second Edition

### Ronald F. Probstein

Department of Mechanical Engineering
Massachusetts Institute of Technology

This book is dedicated with affection to my wife
Irène, whose courage, good humor, and patience
have been an inspiration to me.

$$\eta_r = \frac{\eta}{\mu} = 1 + \left(\frac{2.5\mu_{in} + \mu}{\mu_{in} + \mu}\right)\phi \tag{5.3.24}$$

so for a gas the factor 2.5 in Einstein's equation is replaced by 1.

Einstein's result is remarkable, since it says that for uniform shear the relative viscosity does not depend on the size or size distribution of the spheres but only on the volume fraction, provided the solution is very dilute. A physical explanation for this follows from the diluteness criterion, which may be restated as the interparticle distance being large enough that the motion of any particle is unaffected by that of any neighboring particles. As a result, the increased energy dissipation arising from the presence of the particles must be proportional to the particle number density. Therefore the relative viscosity is simply linear in the particle volume fraction.

One effect neglected in the calculation is the interaction of the particles with the wall; however, this can be shown to be negligible, provided $a/b \ll 1$. A second neglected effect is Brownian motion, which introduces a diffusive flux in addition to the convective viscous flux. So long as the solution is very dilute and the dispersed particles are rigid spheres, the Brownian motion will not alter the mean angular velocity $\dot\gamma/2$, and the Einstein result is unchanged. Although the translational Brownian motion does act on the particle microstructure in trying to uniformize the relative positions of the particles, the relative viscosity is unaffected, since any particle is still unaware of any other particle. The rotational Brownian motion plays no role because of the isotropic behavior of the spherical particles.

If the particles are not spherical, even in the very dilute limit where the translational Brownian motion would still be unimportant, rotational Brownian motion would come into play. This is a consequence of the fact that the rotational motion imparts to the particles a random orientation distribution, whereas in shear-dominated flows nonspherical particles tend toward preferred orientations. Since the excess energy dissipation by an *individual* anisotropic particle depends on its orientation with respect to the flow field, the suspension viscosity must be affected by the relative importance of rotational Brownian forces to viscous forces, although it should still vary linearly with particle volume fraction.

A measure of the importance of Brownian motion is given by the ratio of the Brownian diffusion time to the convection time. The diffusion time may be interpreted as the time taken for a particle to diffuse a distance equal to its radius, which is the characteristic time given by the reciprocal of $D_{rot}$. This time characterizes the time taken for the restoration of the equilibrium micro-structure from a disturbance caused, for example, by viscous convection. The characteristic convection time is simply given by the reciprocal of the shear rate. We denote the ratio of these two times by the Peclet number symbol, since they measure viscous convection to Brownian diffusion, and we write

$$Pe = \frac{\mu\dot\gamma a^3}{kT} \tag{5.3.25}$$

## 5.4 Sedimentation under Gravity

In this section we examine small particles, either solid or fluid, falling (or rising) freely under gravity in a liquid. When the particles are falling, the process is termed *sedimentation*, and when particles are rising, *flotation*. In the former case the particle density is greater than that of the liquid, and in the latter case it is less. We shall generally be concerned with sedimentation, which is used for the separation of dispersed particles from the carrier liquid, for the separation of polydisperse particles in solution according to their size, and for the determination of particle mass. The particle mass is assumed large enough that mass diffusion may be neglected.

For a particle falling freely under gravity, the net force acting on the particle is the difference between the gravitational force and buoyancy force. In Cartesian tensor notation the net force can be written

$$(F_i)_{net} = (\rho - \rho_n)Vg_i \tag{5.4.1}$$

where $\rho$ = particle density
$\rho_n$ = fluid density
$V$ = particle volume

The details of the particle shape are irrelevant, and it does not matter whether the particle continuously turns over and changes in orientation relative to the direction of gravity or whether it moves on a path that is not vertical. For $\rho > \rho_n$ the force will pull the particle down (sedimentation), and for $\rho < \rho_n$ the particle will move up (flotation).

As the particle velocity in a free-fall (or rise) increases, the viscous drag opposing the motion will also increase. For small particles, a steady terminal fall speed is reached very rapidly, in a time of the order of the viscous relaxation time. For a sphere of radius $a$ this time is about $a^2/\nu$ for a density difference of $O(1)$. If the suspending liquid is water, $\nu \approx 10^{-6}$ m$^2$ s$^{-1}$, so even for 100-$\mu$m particles this time is exceedingly short. With small particles the Reynolds numbers are generally sufficiently small that we may neglect inertia and, from Eq. (5.1.3), write for the steady drag force acting on the particle

$$(F_i)_{visc} = -f_{ij}U_j, \tag{5.4.2}$$

Here, $U_i$ is the terminal velocity and $f_{ij}$ is the translational friction tensor; that is, the force depends on the orientation for a particle of arbitrary shape.
With $(F_i)_{net} = -(F_i)_{visc}$,

$$f_{ij}U_j = (\rho - \rho_n)Vg_i \tag{5.4.3}$$

or

$$f_{ij}U_j = m_i\left(1 - \frac{\rho_n}{\rho}\right)g_i \tag{5.4.4}$$

where $n$ is the particle mass. When the translational friction tensor can be replaced by a mean value $\bar{f}$, it follows from Eq. (5.4.4) that the viscous relaxation time $m/f$ for the particle can be determined from a measurement of its fall speed. Note that $m$ alone cannot be determined. This situation is comparable to the classical experiment of J.J. Thomson in which the charge-to-mass ratio of an electron could be determined, but neither by itself.

For a sphere the translational friction coefficient is independent of orientation, and the viscous drag force for a rigid particle of radius $a$ is given by Stokes' drag law, Eq. (5.1.5), with the result

$$6\pi\mu a U = \tfrac{4}{3}\pi a^3(\rho - \rho_{fl})g \qquad (5.4.5)$$

or

$$U = \frac{2}{9}\frac{a^2}{\nu}\left(\frac{\rho}{\rho_{fl}} - 1\right)g \qquad (5.4.6)$$

This shows that the terminal velocity decreases as the square of the decrease in particle size and linearly with a decrease in the density difference. The corresponding Reynolds number based on the particle radius is

$$Re = \frac{Ua}{\nu} = \frac{2}{9}\frac{a^3}{\nu^2}\left(\frac{\rho}{\rho_{fl}} - 1\right)g \qquad (5.4.7)$$

For a density ratio of 2 the Reynolds number will be less than 1 for spherical particles with radii less than about 75 $\mu$m. We are therefore generally justified in neglecting inertia effects for the particle range of interest.

For a liquid drop held spherical by surface tension, the terminal speed from Eq. (5.1.4) is given by

$$U = \frac{1}{3}\frac{a^2}{\nu}\frac{\mu + \mu_{in}}{\mu + \tfrac{3}{2}\mu_{in}}\left(\frac{\rho}{\rho_{fl}} - 1\right)g \qquad (5.4.8)$$

where $\mu_{in}$ is the viscosity of the fluid sphere. The result of Eq. (5.4.6) is recovered with $\mu_{in}/\mu \to \infty$. For a rising spherical gas bubble, where $\mu_{in}/\mu \to 0$ and $\rho/\rho_{fl} \to 0$, the flotation terminal speed is simply $\tfrac{1}{3}a^2 g/\nu$.

Let us consider now a container with an initially homogeneous suspension of particles denser than the liquid with no specification at this stage on the degree of diluteness (Fig. 5.4.1A). If allowed to stand, the particles will settle to the bottom of the container, and at some later time a discrete boundary will be seen separating clarified liquid at the top from the suspension (Fig. 5.4.1B). This boundary will be moving downward. A second discrete boundary will be seen separating the sedimented particles at the bottom from the suspension, and this boundary will be moving upward. After a long enough time all the particles will have sedimented, and an equilibrium state will be reached, as shown in Fig. 5.4.1C.
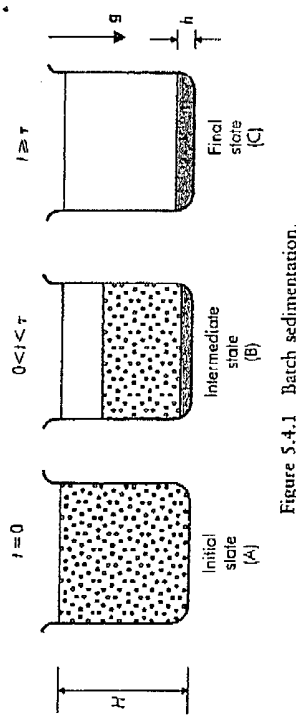
Figure 5.4.1  Batch sedimentation.

The discontinuities diagrammed in Fig. 5.4.1 are termed *kinematic shocks* in that they represent discontinuities in density. Let us calculate the speed at which the top discontinuity moves down and the bottom one up. For specificity consider a downward-moving shock. With respect to a coordinate system moving down with the speed of the discontinuity $u$ (Fig. 5.4.2A), the flow is steady and conservation of mass for the one-dimensional picture considered gives

$$\rho_1(U_1 - u) = \rho_2(U_2 - u) \qquad (5.4.9)$$

Here, $\rho$ is the particle concentration, with the subscript 1 denoting conditions above the discontinuity, and 2 denoting those below. The speed of the shock is therefore

$$u = \frac{j_2 - j_1}{\rho_2 - \rho_1} \qquad (5.4.10)$$

where $j$ is the particle flux passing downward by gravity alone.

In the case of a dilute suspension $U_1 = U_2 = U_0$, where $U_0$ is the infinitely dilute suspension, particle fall speed, which for the case of rigid spheres is given
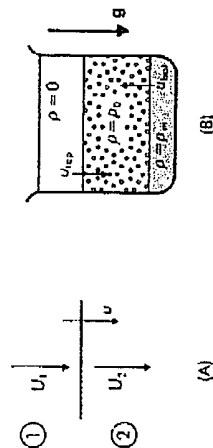


Figure 5.4.2  (A) Kinematic shock; (B) boundary conditions for kinematic shocks in batch sedimentation